

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

M&amp;M-033-USA-PCT

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/719166

INTERNATIONAL APPLICATION NO.  
PCT/JP99/04253INTERNATIONAL FILING DATE  
August 5, 1999PRIORITY DATE CLAIMED  
August 28, 1998TITLE OF INVENTION  
Cationic Photocatalyst Composition And Photocurable Composition

APPLICANT(S) FOR DO/EO/US

Hiroji Fukui

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to promptly begin national examination procedures (35 U.S.C. 371(f)).
4. ☒ The US has been elected by the expiration of 19 months from the priority date (PCT Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☒ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 16 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.  
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information: Claim For Priority

17. ☒ The following fees are submitted:**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :**Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
and International Search Report not prepared by the EPO or JPO ..... \$1000.00International preliminary examination fee (37 CFR 1.482) not paid to  
USPTO but International Search Report prepared by the EPO or JPO ..... \$860.00International preliminary examination fee (37 CFR 1.482) not paid to USPTO but  
international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$710.00International preliminary examination fee paid to USPTO (37 CFR 1.482)  
but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$690.00International preliminary examination fee paid to USPTO (37 CFR 1.482)  
and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$100.00**ENTER APPROPRIATE BASIC FEE AMOUNT =**

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	- 3 - - 20 =	- 0 -	X \$18.00
Independent claims	- 2 - - 3 =	- 0 -	X \$80.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00

\$

**TOTAL OF ABOVE CALCULATIONS =**

\$ 860.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above  
are reduced by 1/2.

\$

**SUBTOTAL =**

\$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

**TOTAL NATIONAL FEE =**

\$ 860.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

+

\$ 40.00

**TOTAL FEES ENCLOSED =**

\$ 900.00

Amount to be  
refunded:

\$

charged:

\$

a. ☒ A check in the amount of \$ 900.00 to cover the above fees is enclosed.b. ☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees.  
A duplicate copy of this sheet is enclosed.c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 20-1424. A duplicate copy of this sheet is enclosed.**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO:

TOWNSEND &amp; BANTA

1225 Eye St., N.W.

Suite 500

Washington, D.C. 20005

phone: (202) 682-4727

SIGNATURE:

Donald E. Townsend

NAME

22,069

REGISTRATION NUMBER

## S P E C I F I C A T I O N

CATIONIC PHOTOCATALYST COMPOSITION AND PHOTOCURABLE  
COMPOSITION5     **TECHNICAL FIELD**

10     The present invention relates to a cationic photocatalyst composition and a photocurable composition incorporating the cationic photocatalyst composition. More particularly, the present invention relates to a cationic photocatalyst composition and a photocurable composition which can assure a sufficient open time after irradiation and enhance adhesive strength after cure.

**BACKGROUND ART**

15     Aryl diazonium, triaryl sulfonium, diaryl iodonium, benzyl sulfonium and phosphonium salts are conventionally known as cationic photocatalysts which polymerize or cure compounds containing an epoxy or other cationically polymerizable group. These cationic photocatalysts are used  
20     in combination with compounds containing cationically polymerizable groups, such as epoxy resins, oxetane derivatives and vinyl ether derivatives, to provide compositions useful for photopolymerization or photocure.

25     A variety of industrial uses have been investigated for the aforementioned compositions utilizing a photocure

reaction, including paints, coatings, varnishes, matrices for composite materials, materials for giving light-developed forms, adhesives and pressure-sensitive adhesives. Particularly, the application of epoxy resins on such various uses has been investigated because they, when cured, show excellent adhesion, creep resistance, light stability, heat stability, chemical stability and the like ("Newly-revised Epoxy Resin", Hiroshi Kakiuchi, Shoko-do, 1985).

In order for compositions to be useful for the above-described purposes, they when cured must show good adhesion to an adherend and good light stability. In addition, they must have the ability to cure quickly when cationically photopolymerized, i.e., fast-curing property. Where they are formulated into paints, coatings or varnishes, curing thereof is desired to complete as soon as irradiation is terminated. That is, it is strongly demanded that curing thereof completes quickly within a short exposure time.

Adhesives and pressure-sensitive adhesives utilizing cationic photopolymerization have been also noted. Such adhesives, when cured via cationic photopolymerization, show excellent levels of creep resistance, light stability, water resistance, heat stability and chemical stability and high adhesive strength. In addition, they show good adhesion to various materials such as metals, plastics and glasses.

Adhesives, pressure-sensitive adhesives or the like have

been thus proposed which utilize cationic photocure of cationically photopolymerizable compounds such as epoxy resins.

When in use for joining adherends, such adhesives, pressure-sensitive adhesives or the like are required to show fast-curing characteristics as required for paints, coatings and varnishes, and to simultaneously provide a sufficiently long work time (a period of time which permits a joint operation after they are irradiated, also referred to as an open time) to permit a joint operation. That is, it is strongly demanded that they remain sufficiently flowable, wet and tacky to permit a joint operation after they have been irradiated.

In Japanese Patent Laying-Open No. Sho 63-248825, slow-curing type UV-curable epoxy resins are disclosed utilizing a fixing agent containing a poly(alkylene oxide) residue portion. The use of fixing agent containing a poly(alkylene oxide) residue portion reduces the occurrence of skinning after exposure to an ultraviolet ray. The skinning refers to a condition wherein a skin layer is caused to cure by irradiation to reduce its wettability, tackiness and the like while an interior maintains its flowability.

In accordance with the teaching of Japanese Patent Laying-Open No. Sho 63-248825, the occurrence of skinning is successfully retarded by the use of fixing agent containing

a poly(alkylene oxide) residue portion. However, since this reference contemplates to satisfy the skinning-preventive property and adhesive property simultaneously by the use of such a fixing agent. It has been accordingly difficult to design such properties independently.

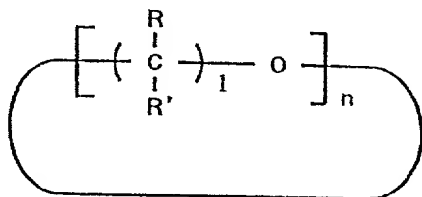
Japanese Patent Laying-Open No. Hei 3-172378 discloses photocurable epoxy adhesive compositions which contain an epoxide cure retarder selected from the group consisting of nitrile- or vinyl ether-substituted organic materials. The use of vinyl ether-substituted organic materials is reported to assure an open time of about 40 minutes. Japanese Patent Laying-Open No. Hei 3-172378 discloses photocurable epoxy adhesive compositions with an extended open time. No reference is however made as to the adhesive property, i.e., adhesive strength of such adhesive compositions.

In view of the current state of the art as described above, it is an object of the present invention to provide a cationic photocatalyst composition and a photocurable composition incorporating the cationic photocatalyst composition which are suitable for use in adhesives or pressure-sensitive adhesives utilizing cationic photopolymerization, which can present a sufficiently long open time after they are irradiated and which, when cured, provide a sufficiently high adhesive strength.

## DISCLOSURE OF THE INVENTION

In accordance with a broad aspect of the present invention, a cationic photocatalyst composition is provided which is characterized as containing a photosensitive onium salt and a compound either represented by the following  
5 formula (1) or having a substituting group represented by the following formula (1).

Formula (1) [Compound B]



In the formula (1), R and R' are suitably selected from hydrogen, halogen, saturated hydrocarbon groups, unsaturated hydrocarbon groups, substituting groups comprised of any  
10 suitable combination of elements such as carbon, hydrogen, oxygen, nitrogen and sulfur, and the like. 1 and n each is an integer of 2 or larger.

In accordance with another broad aspect of the present invention, a photocurable composition is provided which is characterized as containing the above-specified cationic photocatalyst composition and a compound having at least one  
15 cationically polymerizable group in a molecule.

In the present invention, a preferred cationically

polymerizable group is an epoxy group.

A detailed description of the present invention is below given.

The cationic photocatalyst composition in accordance with the present invention contains the above-specified photosensitive onium salt which shows excellent cationic photocatalytic activity, and a compound either represented by the formula (1) or containing, as a substituting group, a structure of the formula (1).

Examples of onium salts include, but not limited to, aromatic diazonium salts, aromatic iodonium salts, aromatic sulfonium salts and the like. The use of photosensitive onium salts which exhibit low thermal catalytic activities in the approximate temperature range of 20 - 80 °C is preferred for their ability to increase the storage stability of the composition.

Specific examples of photosensitive onium salts include IRGACURE 261 (product of Ciba-Geigy), OPTOMER SP-150 (product of Asahi Denka Ind. Co., Ltd.), OPTOMER SP-151 (product of Asahi Denka Ind. Co., Ltd.), OPTOMER SP-170 (product of Asahi Denka Ind. Co., Ltd.), OPTOMER SP-171 (product of Asahi Denka Ind. Co., Ltd.), UVE 1014 (product of General Electronics Co., Ltd.), CD 1012 (product of Sartomer), SANALD SI-60L (product of Sanshin Chem. Ind. Co., Ltd.), SANALD SI-80L (product of Sanshin Chem. Ind. Co.,



Ltd.), SANAID SI-10L (product of Sanshin Chem. Ind. Co.,  
Ltd.), CI-2064 (product of Nippon Soda Co., Ltd.), CI-2639  
(product of Nippon Soda Co., Ltd.), CI-2624 (product of  
Nippon Soda Co., Ltd.), CI-2481 (product of Nippon Soda Co.,  
5 Ltd.), RHODORSIL PHOTOINITIATOR 2074 (product of Rhone  
Poulenc GmbH), UVI-6990 (product of Union Carbide), BBI-103  
(product of Midori Chem. Co., Ltd.), MPI-103 (product of  
Midori Chem. Co., Ltd.), TPS-103 (product of Midori Chem.  
Co., Ltd.), MDS-103 (product of Midori Chem. Co., Ltd.),  
10 DTS-103 (product of Midori Chem. Co., Ltd.), NAT-103  
(product of Midori Chem. Co., Ltd.), NDS-103 (product of  
Midori Chem. Co., Ltd.) and the like.

The above-listed photosensitive onium salts may be used  
alone or in any combination thereof.

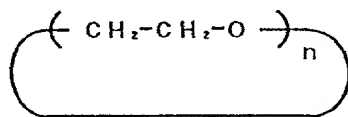
5 The compound either represented by the formula (1) or  
containing, as a substituting group, a structure of the  
formula (1) is incorporated in the composition to extend an  
open time for the case where the cationic photocatalyst  
composition according to the present invention is utilized  
20 for a photocurable composition. As indicated by the formula  
(1), this compound is not particularly specified, so long as  
it is a cyclic compound having ether and alkylene linkages  
or a compound having such a cyclic structure as a  
substituting group. In the above-specified formula (1), R  
25 and R' are not particularly limited and may suitably be

selected from hydrogen, halogen, saturated hydrocarbon groups, unsaturated hydrocarbon groups, substituting groups comprising any suitable combination of elements such as carbon, hydrogen, oxygen, nitrogen and sulfur, and the like.

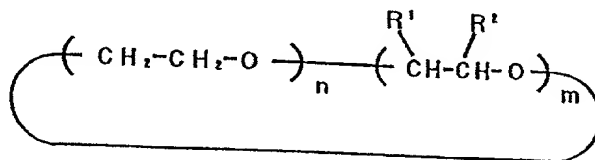
5 1 and n each is an integer of 2 or larger.

The compounds B-1 - B-5 defined as respectively having the following formulas (2) - (6) are preferably used for the compound represented by the formula (1).

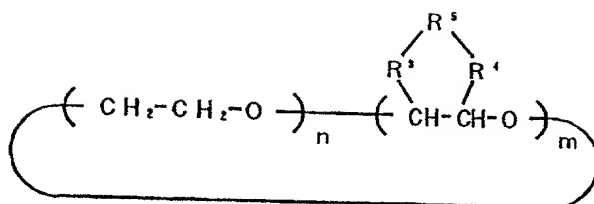
10 Formula (2) [Compound B-1]



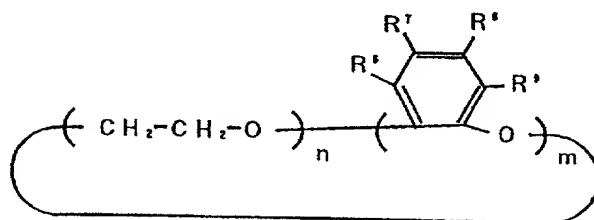
Formula (3) [Compound B-2]



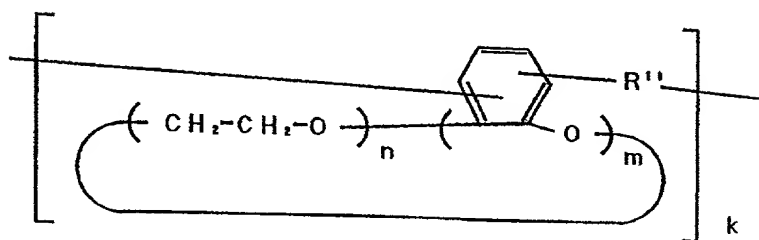
Formula (4) [Compound B-3]



Formula (5) [Compound B-4]



Formula (6) [Compound B-5]



5 In the formulas (3) - (6),  $R^1 - R^{10}$  may be suitably selected from hydrogen, halogen, saturated hydrocarbon groups, unsaturated hydrocarbon groups, substituting groups comprising any suitable combination of elements such as carbon, hydrogen, oxygen, nitrogen and sulfur, and the like.

m, n and k are integers, respectively. While not limiting, those compounds represented by the formulas (3) - (6) may have their respective repeating units arranged in a block or random fashion.

5           Specific examples of compounds represented by the formula (1) include 12-crown-4, 15-crown-5, 18-crown-6, 24-crown-8, 30-crown-10, 2-aminomethyl-12-crown-4, 2-aminomethyl-15-crown-5, 2-aminomethyl-18-crown-6, 2-hydroxymethyl-12-crown-4, 2-hydroxymethyl-15-crown-5, 2-hydroxymethyl-18-crown-6, dicyclohexano-18-crown-6, dicyclohexano-24-crown-8, dibenzo-18-crown-6, dibenzo-24-crown-8, dibenzo-30-crown-10, benzo-12-crown-4, benzo-15-crown-5, benzo-18-crown-6, 4'-aminobenzo-15-crown-5, 4'-bromobenzo-15-crown-5, 4'-formylbenzo-15-crown-5, 4'-nitrobenzo-15-crown-5, bis[(benzo-15-crown-5)-15-ylmethyl] pimelate, poly[(dibenzo-18-crown-6)-coformaldehyde], and the like.

10           The compounds either represented by the formula (1) or containing a structure of the formula (1) as a substituting group may be used alone or in any combination.

20           Preferably, the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group is incorporated into the cationic photocatalyst composition of the present invention in the amount of 0.001 - 100 parts by weight, based on 1 part by

25

weight of the photosensitive onium salt. The loading of the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group in the amount of below 0.001 parts by weight may result in the failure to fully obtain an effect of extending an open time as a result of its addition. On the other hand, the loading thereof in the amount of above 100 parts by weight may extend an open time but slow a cure reaction to the level substantially close to termination, resulting in the difficulty to simultaneously optimize the open time and cure rate. More preferably, the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group is incorporated into the cationic photocatalyst composition of the present invention in the amount of 0.01 - 10 parts by weight, based on 1 part by weight of the photosensitive onium salt. This facilitates the simultaneous optimization of open time and cure rate.

Besides the above-described onium salt and the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group, the cationic photocatalyst composition in accordance with the present invention may further contain suitable additives, such as a reactive diluent, a solvent and the like.

The photocurable composition in accordance with the present invention is characterized as containing the

cationic photocatalyst composition in accordance with the present invention and a compound containing at least one cationically polymerizable group in a molecule.

The cationically polymerizable group incorporated in the compound is not particularly specified in type, so long as it is a functional group that polymerizes by a cationic mechanism, and may be vinyloxy, styryl, epoxy, oxetanyl or the like. An epoxy group is preferred for its ability to provide good adhesion and durability.

Specific examples of vinyloxy-containing compounds include, but not limited to, n-propyl vinyl ether, n-butyl vinyl ether, isobutyl vinyl ether, tert-butyl vinyl ether, tert-amyl vinyl ether, cyclohexyl vinyl ether, 2-ethylhexyl vinyl ether, dodecyl vinyl ether, octadecyl vinyl ether, 2-chloroethyl vinyl ether, ethylene glycol butyl vinyl ether, triethylene glycol methyl vinyl ether, 4-vinyloxy butyl benzoate, ethylene glycol divinyl ether, diethylene glycol divinyl ether, triethylene glycol divinyl ether, tetraethylene glycol divinyl ether, butane-1,4-diol-divinyl ether, hexane-1,6-diol-divinyl ether, cyclohexane-1,4-dimethanol-divinyl ether, di(4-vinyloxy) butyl isophthalate, di(4-vinyloxy) butyl glutarate, di(4-vinyloxy) butyl succinate, trimethylolpropane trivinyl ether, 2-hydroxyethyl vinyl ether, 4-hydroxybutyl vinyl ether, 6-hydroxyhexyl vinyl ether, cyclohexane-1,4-dimethanol-monovinyl ether,

diethylene glycol monovinyl ether, 3-aminopropyl vinyl ether, 2-(N,N-diethylamino)ethyl vinyl ether, urethane vinyl ether, polyester vinyl ether and the like.

5 Examples of styryl-containing compounds include, but not limited to, styrene, p-methylstyrene,  $\alpha$ -methylstyrene, p-methoxystyrene, p-tert-butoxystyrene, p-chloromethylstyrene, p-acetoxystyrene, divinylbenzene and the like.

10 Examples of epoxy-containing compounds include, but not limited to, bisphenol A based epoxy resins, hydrogenated bisphenol A based epoxy resins, nobolac type epoxy resins, cycloaliphatic epoxy resins, brominated epoxy resins, rubber modified epoxy resins, urethane modified epoxy resins, glycidyl ester compounds, epoxidized polybutadiene, epoxidized styrene-butadiene-styrene and the like.

15 The above-listed compounds having at least one cationically polymerizable group in a molecule may be used alone or in any combination. Other applicable compounds may have two different types of cationically polymerizable groups in a molecule.

20 Preferably, the photocurable composition in accordance with the present invention contains the aforementioned compound having a cationically polymerizable group in the amount of 10 - 10,000 parts by weight, based on 1 part by weight of the aforementioned photosensitive onium salt. If  
25 the loading of compound having a cationically polymerizable

group falls below 10 parts by weight, its curing may become insufficient to provide mechanical strength, possibly resulting in the failure to provide expected adhesion and durability. On the other hand, if its loading goes beyond 10,000 parts by weight, a loading proportion of the photosensitive onium salt, while effective to cause the photocurable composition to cure, becomes relatively small, possibly resulting in the difficulty to increase a cure rate to a sufficient level even if a loading proportion of the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group is caused to decrease.

For the aforesaid reasons, it is more preferred that the photocurable composition contains 0.001 - 100 parts by weight of the compound either represented by the formula (1) or containing, as a substituting group, a structure of the formula (1), and 10 - 10,000 parts by weight of the compound having a cationically polymerizable group, respectively based on 1 part by weight of the photosensitive onium salt.

When desired, a sensitizer may be added to promote photopolymerization or photocure. Any sensitizer can be used, so long as it serves to increase a radiation sensitivity of the composition. Examples of sensitizers include anthracene, perylene, coronene, tetracene, benzanthracene, phenothiazine, flavin, acridine,



ketocoumarin, thioxanthone derivatives, benzophenone, acetophenone and the like.

Preferred for use as the sensitizer are thioxanthone derivatives which exhibit the excellent ability to increase a sensitivity of the composition and can be handled easily.

Examples of thioxanthone derivatives include, but not limited to, 2-chlorothioxanthone, 2,4-dimethylthioxanthone, 2,4-diethylthioxanthone, 2,4-diisopropylthioxanthone, isopropylthioxanthone and the like.

When present, the amount of thioxanthone derivative sensitizer is preferably in the range of 0.01 - 100 parts by weight, based on 1 part by weight of the aforementioned photosensitive onium salt. The loading of thioxanthone derivative sensitizer in the amount of below 0.01 parts by weight may result in the failure to obtain a sufficient sensitizing action to promote a cure rate. On the other hand, if the loading of thioxanthone derivative sensitizer exceeds 100 parts by weight, its sensitizing action may become excessive to result in the difficulty to assure a desirable open time.

For the aforesated reasons, it is more preferred that the photocurable composition contains 0.001 - 100 parts by weight of the compound either represented by the formula (1) or containing, as a substituting group, a structure of the formula (1), 10 - 10,000 parts by weight of the compound

having a cationically polymerizable group and 0.01 - 100 parts by weight of thioxanthone derivative sensitizer, respectively based on 1 part by weight of the photosensitive onium salt.

5 Preferably, the cationic photocatalyst composition and photocurable composition in accordance with the present invention are photosensitive in the 300 - 800 wavelength region of the spectrum, wherein at least photosensitive onium salt and thioxanthone derivative are photosensitized.

10 If photosensitive only in the region below 300 nm, the thick application of photocurable composition may result not only in the increased occurrence of skinning at an incident surface, but also in the failure to achieve uniform curing of the applied composition from its surface layer to deep interior. On the other hand, if photosensitive only in the  
15 region above 800 nm, it may become difficult to receive a sufficient radiation energy to accelerate a cure rate, while possible to reduce the occurrence of skinning and achieve uniform curing from the surface layer to deep interior. It  
20 is accordingly desired that the cationic photocatalyst composition and photocurable composition be photosensitive in the 300 - 800 nm wavelength region of the spectrum, as stated above.

25 A radiation source used to irradiate the photocatalyst or photocurable composition in accordance with the present

invention is not particularly specified in type, but preferably has an emission distribution in the 300 - 800 nm region. Suitable sources of radiation include low-pressure mercury lamps, medium-pressure mercury lamps, high-pressure mercury lamps, ultrahigh-pressure mercury lamps, chemical lamps, blacklight lamps, microwave-excited mercury lamps, metal halide lamps, sodium lamps, fluorescent lamps and the like.

For the purposes of suppressing curing in a surface layer only and achieving internal curing of the applied composition, such radiation sources may be used in such a controlled fashion that prevents the passage of radiation in the region of below 300 nm. A natural light, such as sunlight, may also be used.

The photocurable composition in accordance with the present invention is suitable for use in the constitution of adhesives, molded products and the like. Suitable uses include hot melt adhesives, adhesive sheets, liquid adhesives, casting compositions and the like.

In the case where the photocurable composition in accordance with the present invention is used as a hot melt adhesive, the photocurable composition is desired to melt in the 50 - 200 °C temperature range and has a viscosity at 200 °C of not exceeding 1,000,000 cps. If the photocurable composition has a property of melting at a temperature of

below 50 °C, its cohesive strength at normal temperature may be lowered to result in the failure to exhibit an expected initial adhesion immediately after application. On the other hand, if having a property of starting to melt at a temperature of above 200 °C, the composition when melt at a high temperature may cause an adherend to undergo thermal deterioration. If the viscosity at 200 °C of photocurable composition exceeds 1,000,000 cps, its wettability and flowability may be markedly reduced to possibly result in the difficulties to perform its application to an adherend and insure a sufficient initial adhesive strength.

Where the photocurable composition in accordance with the present invention is used as a hot melt adhesive, it preferably shows a softening point in the 30 - 180 °C range when measured according to a ring and ball method. If the photocurable composition has a softening point of below 30 °C, its cohesive strength at normal temperature may be lowered to result in the failure to exhibit an expected initial adhesion immediately after it is applied. On the other hand, if having a softening point of above 180 °C, the photocurable composition when melt at a high temperature may cause an adherend to undergo thermal deterioration.

In the case where the photocurable composition in accordance with the present invention is used as a hot melt adhesive to join adherends together, the following procedure

may preferably be utilized: The photocurable composition is allowed to melt thermally, applied, in a molten state, onto one or both of the adherends, exposed to a radiation either before or after the application, followed by joining the adherends together.

A photocurable adhesive sheet can be obtained by sheeting the photocurable composition in accordance with the present invention. Preferably, this photocurable adhesive sheet, prior to being cured, has a dynamic storage shear modulus of  $10^3$  Pa -  $10^{10}$  Pa at a frequency of 10 Hz at a temperature of 0 - 50 °C. If the dynamic storage shear modulus is below  $10^3$  Pa, the cohesive strength of photocurable composition may be lowered to result in the difficulty to retain its original sheet form. On the other hand, if the dynamic storage shear modulus exceeds  $10^{10}$  Pa, the cohesive strength of photocurable composition may be caused to increase to a significant degree that renders it less wettable or adherent with respect to an adherend, possibly resulting in the failure to obtain a sufficient adhesive strength.

The photocurable composition in accordance with the present invention can also be used as a liquid adhesive, as stated earlier. In such an instance, the photocurable composition preferably exhibits a viscosity of 1 - 1,000,000 cps in the 0 - 40 °C temperature range. If the viscosity

falls below 1 cps, the photocurable composition may become excessively flowable to move readily to an area that is not desired to be applied, resulting in the reduced workability. On the other hand, the viscosity of above 1,000,000 cps may lead to a marked reduction in flowability of the composition to result in the difficulty to perform a coating operation.

A technique used to join members using the photocurable composition or photocurable adhesive sheet in accordance with the present invention can be suitably chosen depending upon the configurations of photocurable composition or photocurable adhesive sheet and others.

When the aforementioned photocurable composition is used as an adhesive to join adherends together, the photocurable composition is applied onto one or both of the adherends and then exposed to radiation, followed by placing the adherends in contact with each other. In this case, due to the incorporation of compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group, the photocurable composition when irradiated assures a sufficient open time to perform a joint operation.

When the photocurable adhesive sheet is used to join a pair of adherends together, the photocurable adhesive sheet may be exposed to radiation either before or after those adherends are brought into contact with each other. Since

the incorporation of compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group assures a sufficient open time, it is preferred to utilize a procedure whereby the photocurable adhesive sheet is applied to one of the adherends and subsequently exposed to radiation, followed by joining the adherends together by the intervening photocurable adhesive sheet.

In addition to being useful for adhesives, the photocurable composition in accordance with the present invention can be used as a casting composition. In such an instance, the photocurable composition preferably exhibits a viscosity at 20 °C of not exceeding 1,000,000 cps. If its viscosity exceeds 1,000,000 cps, the photocurable composition becomes less flowable to possibly result in the difficulty to cast it into a mold.

Illustrating one preferred technique used to obtain a molded product, the photocurable composition is exposed to a light and then introduced into a mold where it is allowed to cure. Also in this case, the photocurable composition can be introduced smoothly into a mold due to the inclusion of the compound either represented by the formula (1) or containing, as a substituting group, a structure of the formula (1), which acts to retard a cure rate. If the photocurable composition is irradiated after it is cast into

a mold, uneven exposure thereof to a light may occur to result in the difficulty to obtain a homogeneous product. On the other hand, irradiating the photocurable composition before it is cast into a mold, as described above, allows even exposure thereof to a light, resulting in obtaining a homogeneous product, i.e., a molded product having a narrow variation in cure level or in mechanical strength throughout in the product.

Suitable additives, such as tackifiers, thickeners and extenders generally known in the art, can be added to the cationic photocatalyst or photocurable composition in accordance with the present invention within the range that is not detrimental to the purposes of this invention.

Examples of tackifiers include rosin resins, modified rosin resins, terpene resins, terpene-phenolic resins, aromatic modified terpene resins, C5 or C9 petroleum resins, coumarone resins and the like. Particularly when adherends consist of polyolefins, the use of rosin or petroleum resins is preferred for their ability to build strong adhesion.

Other additives can also be added to improve coating properties, examples of which include thickeners such as acrylic, epichlorohydrin, isoprene and butyl rubbers; thixotropic agents such as colloidal silica and polyvinyl pyrrolidone; extenders such as calcium carbonate, titanium oxide and clay; and adjusting agents such as polyester,



acrylic polymer, polyurethane, silicone, polyether,  
polyvinyl ether, polyvinyl chloride, polyvinyl acetate,  
polyisobutylene and waxes.

Where the photocurable composition in accordance with  
the present invention is utilized as an adhesive, suitable  
materials can be added to improve its shear bond strength,  
examples of which include inorganic balloons as comprised of  
glass, alumina and ceramic; organic spheres such as nylon,  
acrylic and silicon beads; organic balloons as comprised of  
vinylidene chloride and acrylics; and single-component  
fibers such as comprised of glass, polyester, rayon, nylon,  
cellulose and acetate.

The glass fibers can be added, in the form of fibrous  
chips, to the composition. The improved shear bond strength  
is obtained if the photocurable composition is polymerized  
in the form of being impregnated in a glass woven fabric.

(ACTION)

The cationic photocatalyst composition in accordance  
with the present invention contains the compound either  
represented by the formula (1) or containing a structure of  
the formula (1) as a substituting group, in addition to the  
photosensitive onium salt. The compound either represented  
by the formula (1) or containing a structure of the formula  
(1) as a substituting group, if mixed with a cationically  
photopolymerizable compound, acts to retard a cure rate and

assure a sufficient open time after the mixture is exposed to radiation. This is probably because the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group is a compound that  
5 contains cyclic ether.

While acting to retard a cure rate, the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group does not affect the adhesive strength after it is cured. The presence of such a  
10 compound thus serves to impart a sufficient open time and a sufficient level of adhesive strength to the photocurable composition after it is cured.

Accordingly, a sufficient length of open time and a sufficient level of adhesive strength can be satisfied  
15 simultaneously and designed independently of each other.

Due to the inclusion of the above-described cationic photocatalyst composition and compound having a cationically polymerizable group, the photocurable composition according to the present invention not only assures a sufficient open  
20 time but also provides a sufficient adhesive strength after it is cured.

Where the cationically polymerizable group is an epoxy, the further increased adhesion and durability result.

The further inclusion of a thioxanthone derivative  
25 increases a cure rate as a result of its sensitizing action.

That is, an open time and cure rate can be easily controlled by suitably adjusting a blending ratio of the thioxanthone derivative to the compound either represented by the formula (1) or containing a structure of the formula (1) as a substituting group.

#### BEST MODE FOR CARRYING OUT THE INVENTION

The following non-limiting Examples illustrate the present invention more specifically.

(EXAMPLE 1 - Preparation of a Cationic Photocatalyst Composition)

20 g of a photosensitive onium salt, OPTOMER SP170 of Asahi Denka Ind. Co., Ltd., and 10 g of 18-crown-6 as a compound represented by the formula (1), were charged into a sample bottle having a volume of about 50 ml where they were mixed and agitated by a stirrer to obtain a cationic photocatalyst composition. 18-crown-6 represents a case of n=6 for the compound B-1 having the structure as specified above.

(EXAMPLE 2 - Preparation of a Cationic Photocatalyst Composition)

20 g of a photosensitive onium salt, OPTOMER SP150 of Asahi Denka Ind. Co., Ltd., and 10 g of 18-crown-6, as a compound represented by the formula (1), were charged into a sample bottle having a volume of about 50 ml where they were

mixed and agitated by a stirrer to obtain a cationic photocatalyst composition.

(EXAMPLE 3 - Preparation of a Cationic Photocatalyst Composition)

5        20 g of a photosensitive onium salt, UVI-6990 of Union Carbide Corp., and 10 g of 18-crown-6, as a compound represented by the formula (1), were charged into a sample bottle having a volume of about 50 ml where they were mixed and agitated by a stirrer to obtain a cationic photocatalyst composition.

(EXAMPLE 4 - Preparation of a Photocurable Composition)

10        9 g of the cationic photocatalyst composition obtained in Example 1, and 300 g of a bisphenol A based epoxy resin (manufactured by Yuka-Shell Epoxy Co., Ltd., product name: EPICOAT 823), as an epoxy resin compound, were charged into a 0.5 liter separable flask where they were mixed and agitated to obtain a photocurable composition. The photocurable composition such obtained revealed a viscosity of 14,000 cps at 25 °C.

15        The photocurable composition obtained in the manner as described above was used as a liquid adhesive composition to evaluate a period of time the adhesive composition remained unaccompanied by the skinning after it had been irradiated, as well as its shear bond strength, according to the below-described evaluation methods. As a result, the skinning

20

25

appeared 90 minutes after it had been irradiated. The shear bond strength reached 110 kgf/cm<sup>2</sup>. When the stainless steel plates were placed in contact with each other 5 minutes after the adhesive composition had been irradiated, the shear bond strength reached 103 kgf/cm<sup>2</sup>.

(COMPARATIVE EXAMPLE 1)

The procedure of Example 4 was followed, except that 18-crown-6, as the compound represented by the formula (1), was not used, to obtain a photocurable composition. This photocurable composition revealed a viscosity of 14,000 cps at 25 °C.

The photocurable composition such obtained was used as a liquid adhesive composition to evaluate its properties in the same manner as in Example 4. As a result, skinning occurred within a minute after the adhesive composition had been irradiated. The shear bond strength reached 110 kgf/cm<sup>2</sup> when stainless steel plates were placed in contact with each other immediately after the adhesive composition had been irradiated, but a value of below 1 kgf/cm<sup>2</sup> when they were placed in contact with each other 5 minutes after the adhesive composition had been irradiated.

As can be appreciated from a comparison between Example 4 and Comparative Example 1, the adhesive composition of Example 4, because of its incorporation of the compound represented by the formula (1), exhibits a marked extension

of the time until skinning occurs, as well as the increased shear bond strength when stainless steel plates are placed in contact with each other 5 minutes after it is irradiated, i.e., the sufficiently increased cure rate.

5 (EXAMPLES 5 - 8)

The procedure of Example 4 was repeated, except that the photosensitive onium salt, compound represented by the formula (1) and epoxy compound, respectively specified in Table 1, were blended in the ratios (by weight) indicated in  
10 Table 1, to provide photocurable compositions which were subsequently evaluated in the same manner as in Example 4. The results are given in Table 1. Dibenzo 18-crown-6 represents a case of  $n=6$ ,  $m=2$  and  $R^6 - R^9 = \text{hydrogen}$  for the compound B-6 having the structure as specified above.

15 (COMPARATIVE EXAMPLES 2 - 5)

The procedure of Example 4 was repeated, except that the compound represented by the formula (1) was excluded and the photosensitive onium salt and epoxy compound, each specified in Table 1, were blended in the ratios (by weight)  
20 indicated in Table 1, to provide photocurable compositions of Comparative Examples 2 - 5 which were subsequently evaluated in the same manner as in Example 4. The results are given in Table 1.

As apparent from Table 1, in the case of Examples 4 - 8  
25 where the photocurable compositions contain the compound

represented by the formula (1) as well as the photosensitive onium salt, a period of time each photocurable composition remains unaccompanied by skinning after it has been irradiated is extended so that an adhesion operation can be eased.

In contrast, for the photocurable compositions obtained in Comparative Examples 1 - 5, respectively corresponding to those obtained in Examples 4 - 8, skinning appeared soon after they had been irradiated. This demonstrates that the photocurable compositions obtained in Examples 4 - 8, when used as adhesives, provide a sufficient time to carry out a joint operation, while those obtained in Comparative Examples 1 - 5 must be subjected to a joint operation soon after irradiation in order to obtain sufficient adhesive strength.

(EXAMPLE 9)

9 parts by weight of the cationic photocatalyst composition obtained in Example 1, 120 parts by weight of an alicyclic epoxy resin (product name: ERL4221, manufactured by Union Carbide Corp.), 120 parts by weight of epoxidized SBR (product name: EPOFRIEND A1020, manufactured by Daicel Chem. Ind. Co., Ltd.) and 60 parts by weight of a tackifier (unhydrogenated oil resin manufactured by Mitsui Petrochem. Ind. Co., Ltd., product name: FTR6125) were charged into a planetary mixer equipped with a jacket for circulating a hot

oil. After the mixer was entirely covered with an aluminum foil, the contents were mixed at 30 rpm at 150 °C for 30 minutes to obtain a photocurable composition. This composition revealed a viscosity of 7,000 cps at 150 °C and a softening point of 90 °C.

The photocurable composition obtained in the manner as described above was used as a reactive hot melt adhesive to evaluate an open time and peel adhesion according to the below-described evaluation methods. The measured open time was 5 minutes.

The peel adhesion reached 2.5 kgf/25 mm when stainless steel plates were brought into contact with each other immediately after the adhesive had been irradiated, and 2.3 kgf/25 mm when they were placed in contact with each other 5 minutes after the adhesive had been irradiated.

#### (COMPARATIVE EXAMPLE 6)

The procedure of Example 9 was followed, except that the compound represented by the formula (1) was excluded, to obtain a photocurable composition. This photocurable composition revealed a viscosity of 7,000 cps at 150 °C and a softening point of 90 °C.

The above-obtained photocurable composition was used as a reactive hot melt adhesive, as similar to Example 9, to evaluate an open time and peel adhesion. As a result, the open time was found to be less than 1 minute. The peel



adhesion reached 2.5 kgf/25 mm when stainless steel plates were placed in contact with each other immediately after the adhesive had been irradiated. Joining thereof was found difficult when they were placed in contact with each other 5 minutes after the adhesive had been irradiated.

(EXAMPLE 10 AND COMPARATIVE EXAMPLE 7)

The procedure of Example 9 was repeated, except that the ingredients were blended in the proportions indicated in Table 2, to obtain photocurable compositions of Example 10 and Comparative Example 7.

The photocurable compositions such obtained were evaluated in the same manner as in Example 9 for viscosity, softening point, open time and peel adhesion. The results are given in Table 2.

As can be seen from the shown results, the photocurable compositions obtained in Examples 9 and 10, because of their inclusion of the compound represented by the formula (1), provide the sufficiently increased open time to carry out a joint operation after they have been irradiated, compared to those obtained in Comparative Examples 6 and 7. On the other hand, the photocurable compositions obtained in the corresponding Comparative Examples 6 and 7 must be subjected to a joint operation immediately after irradiation in order to obtain sufficient adhesive strength.

(EXAMPLE 11)

100 g of the photocurable composition obtained in Example 9, and 100 g of a liquid mixture of ethyl acetate and toluene (in a 70/30 ratio by weight) were charged into a 0.5 liter separable flask where they were mixed to provide a solution containing the photocurable composition. This solution was subsequently coated onto a release surface of a polyethylene terephthalate film by a Baker applicator and dried at 110 °C for 5 minutes to provide a 100 µm thick, photocurable adhesive sheet. The resulting photocurable adhesive sheet was found to have a dynamic storage shear modulus of  $10^6$  at a frequency of 10 Hz at a temperature of 25 °C.

The photocurable adhesive sheet such obtained was evaluated for open time and peel adhesion according to the below-described methods. As a result, the measured open time was 5 minutes. The peel adhesion reached 2.5 kgf/25 mm when stainless steel plates were placed in contact with each other immediately after the adhesive had been irradiated, and 2.3 kgf/25 mm when they were placed in contact with each other 5 minutes after the adhesive had been irradiated.

(COMPARATIVE EXAMPLE 8)

The procedure of Example 11 was followed, except that the composition of Comparative Example 6 was replaced by the composition of Example 9, to obtain a photocurable adhesive sheet (100 µm thick). The resulting photocurable adhesive

sheet was found to have a dynamic storage shear modulus of  $10^6$  at a frequency of 10 Hz at a temperature of 25 °C.

The photocurable adhesive sheet was evaluated in the same manner as in Example 11. As a result, the open time was found to be less than 1 minute. The peel adhesion reached 2.5 kgf/25 mm when stainless steel plates were placed in contact with each other immediately after the adhesive had been irradiated. Joining thereof failed when they were placed in contact with each other 5 minutes after the adhesive had been irradiated.

(EXAMPLE 12)

The photocurable composition obtained in Example 5 was filled in a 1,000 mL syringe. A polyvinyl chloride tube was then connected to a discharge port of the syringe. The photocurable composition was allowed to flow through the polyvinyl chloride tube, while exposed to radiation such that a radiation having a wavelength of 365 nm provided an exposure energy of 900 mJ/cm<sup>2</sup>, into a molding tool (made of Teflon, 100 mm x 100 mm x 110 mm). The photocurable composition poured into the molding tool was caused to cure at 25 °C for 7 days, and thereafter taken out from the molding tool to obtain a molded product having a shape of regular parallelepiped.

(COMPARATIVE EXAMPLE 9)

In the attempt to obtain a molded product, the

procedure of Example 12 was followed, with the exception that the composition of Example 5 was replaced by the composition obtained in Comparative Example 2. This attempt however failed because the photocurable composition was thickened during its passage through the polyvinyl chloride tube to the degree that prevented its flow into the molding tool.

#### (EVALUATION METHODS)

In Examples 1 - 12 and Comparative Examples 1 - 9, the following test methods were utilized to evaluate (1) a period of time the composition remained unaccompanied by skinning after it had been irradiated, (2) shear bond strength, (3) open time and (4) peel adhesion.

##### (1) Time Measurements

The procedure used to measure a period of time each photocurable composition remains unaccompanied by skinning after it has been irradiated follows:

The photocurable composition is coated on a stainless steel (SUS 304) plate to a thickness of 50  $\mu\text{m}$ , and then exposed to 30  $\text{mW}/\text{cm}^2$  of radiation concentrated around 365 nm for 30 seconds. The occurrence of skinning at an exposed surface is inspected by touching it with a finger. That is, when the exposed surface finally provides a wet- or tack-free touch, skinning is judged to have occurred and the time elapsed is recorded as a period of time the photocurable

composition remains unaccompanied by skinning after it has been irradiated.

## (2) Shear Bond Strength

Accoring to JIS Z 6850, a 150 mm long and 2 mm thick stainless steel (SUS304) plate (may also be referred to as an adherend A) is abraded at its surface with a wet abrasive paper, # 280, the surface is degreased with ethyl acetate and then dried, and the photocurable composition is coated on the surface to a thickness of 50  $\mu\text{m}$ . Subsequently, the coated composition is irradiated with a radiation source in the region of 300 - 370 nm such that a radiation having a wavelength of 365 nm provides an intensity of 30  $\text{mW}/\text{cm}^2$  for 30 seconds. Either immediately or specified periods of time after the composition has been irradiated, the adherend A coated with the composition is placed in contact with a separately-prepared second adherend A to provide pieces for shear bond strength test. After being left to cure at 25  $^{\circ}\text{C}$  for 7 days, each test piece is subjected to shear bond strength measurement at a pulling rate of 10 mm/min by a tensile tester.

## (3) Open Time

In the assembly of adhesion test pieces in (2) or (4), the composition coated on the adherend is irradiated and, after specified periods of time, the adherend is brought into contact with the second adherend. A maximum period of

time that permits the assembly of the test piece showing an adhesive strength about comparable to that of the test piece assembled immediately after irradiation is defined as an open time.

5 (4) Peel Adhesion

The composition obtained in Example 9, Example 10, Comparative Example 6 or Comparative Example 7 is coated on an SPCC dull steel plate (25 x 125 x 1.6 mm) by a roll coater set at 150 °C to a thickness of 50 - 150 µm. The coating of each composition is exposed to a radiation source in the region of 300 - 370 nm such that a radiation having a wavelength of 365 nm provides an intensity of 30 mW/cm<sup>2</sup> for 30 seconds.

10 In the case of the composition obtained in Example 9 or Comparative Example 6, either immediately or specified periods of time after it has been irradiated, a number 9 cotton fabric (25 x 125 mm) is placed thereon and pressed at a pressure of 0.5 kgf/cm<sup>2</sup> at 23 °C for 2 minutes to obtain a piece for peel adhesion test. After being left to cure at 20 25 °C for 7 days, each test piece is subjected to a dancer roll peeling test according to JIS K 6854 to measure its peel adhesion.

25 In the case of the composition obtained in Example 10 or Comparative Example 7, either immediately or specified periods of time after it has been irradiated, a number 9

cotton fabric (25 x 125 mm) is placed thereon, heat pressed at a pressure of 0.5 kgf/cm<sup>2</sup> at 120 °C for 2 minutes and pressed again at a pressure of 0.5 kgf/cm<sup>2</sup> at 120 °C for 2 minutes to obtain a piece for peel adhesion test. After  
5 being left to cure at 25 °C for 7 days, each test piece is subjected to a dancer roll peeling test according to JIS K 6854 to measure its peel adhesion.

[TABLE 1]

Photo-curable Composition	Cationic Photo-catalyst Composition	Compound (A) (P.B.W.)	Compound (B) (P.B.W.)	Compound (C) (P.B.W.)	Ex.	Comp. Ex.	Ex.	Comp. Ex.	Ex.	Comp. Ex.	Ex.	Comp. Ex.	Ex.	Comp. Ex.
					4	1	5	2	6	3	7	4	8	5
Evaluation Items	Viscosity (At 25°C) (CPS)	SP-170	SP-150	18-CROWN-6	2	2	2	2	2	2	2	2	2	2
		DIBENZO-18-CROWN-6	EPCOAT828	EP4080	1		1		1		1		1	
		CHDVE	EP4080	CHDVE			100	100	100	100	100	100	100	100
	Time Measure (Min.)	Jointed IMM. After Irradiation	Jointed 5Min. After Irradiation	Jointed 5Min. After Irradiation	14000	14000	3500	3500	5	5	3500	3500	3500	3500
					90	<1	60	<1	10	<1	40	<1	90	<1
					110	113	108	105	54	58	105	107	109	105
					103	<1	106	<1	49	<1	106	<1	102	<1



In Table 1, the details of the commercial products used are as follows.

EPICOAT 828: bisphenol A based epoxy resin (Yuka-Shell epoxy resin)

5 EP-4080: hydrogenated bisphenol A epoxy resin (product of Asahi Denka Co., Ltd.)

CHDVE: cyclohexanedimethanol divinyl ether

[TABLE 2]

				Ex.	Comp. Ex.	Ex.	Comp. Ex.
				9	6	10	7
Photo- curable Compo- sition	Cationic Photo- catalyst Compo- sition	Compound (A) (P.B.W.)	SP-170	2	2	2	2
		Compound (B) (P.B.W.)	18- CROWN-6	1		1	
	Compound (C) (P.B.W.)		ERL4221	40	40		
			EPO- FRIEND A 1020	40	40	60	60
			EPICOAT RXE 21			40	40
	TACKIFIER FTR6125			20	20		
15 Evalu- tion Items	Viscosity(At 150°C) (CPS)			7000	7000	510000	510000
	SOFTENING POINT (°C)			90	90	90	90
	OPEN TIME (Min.)			5	<1	5	<1
	Peel Adhesion (kgf/mm)	Jointed IMM. After Irradiation		2.5	2.5	2.3	2.1
		Jointed 5Min. After Irradiation		2.3		2.2	

In Table 2, the details of the commercial products used are as follows.

ERL 4221: alicyclic epoxy resin (manufactured by Union Carbide Japan Co., Ltd.)

5 EPOFRIEND: epoxidized SBS (manufactured by Daicel Chem. Ind. Co., Ltd.)

EPICOAT RXE21: aliphatic epoxy resin (Yuka-Shell epoxy resin)

10 FTR 6125: unhydrogenated petroleum resin (manufactured by Mitsui Petrochem. Co., Ltd.)

#### EFFECTS OF THE INVENTION

15 The cationic photocatalyst composition in accordance with the present invention contains the compound represented by the formula (1) relative to a photosensitive onium salt as a cationically photopolymerizable catalyst. When such a composition is used in combination with a cationically photopolymerizable compound and irradiated, the action of the compound represented by the formula (1) not only insures  
20 a sufficient open time after irradiation, but also results in the provision of a cured product which exhibits the same level of adhesion as those obtained by using conventional cationic photocatalysts.

25 Accordingly, the photocurable composition in accordance with the presents invention, when irradiated, presents a

sufficient open time that allows an easy joint operation and when cured, provides a sufficient adhesive strength.

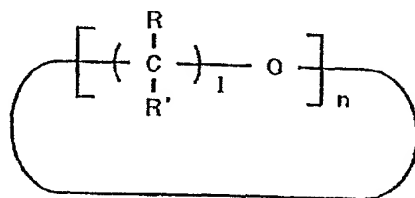
Also, where the cationically photopolymerizable group is epoxy, the composition when cured provides further

5 enhanced adhesion and durability.

# CLAIMS

1. A cationic photocatalyst composition characterized as containing a photosensitive onium salt and a compound  
 5 either represented by the following formula (1) or containing, as a substituting group, a structure of the following formula (1);

(1) [Compound B]



10 (wherein R and R' are suitably selected from hydrogen, halogen, saturated hydrocarbon groups, unsaturated hydrocarbon groups, substituting groups comprising any suitable combination of elements such as carbon, hydrogen, oxygen, nitrogen and sulfur; and 1 and n each is an integer  
 15 of 2 or larger).

2. A photocurable composition characterized as containing:

the cationic photocatalyst composition as recited in claim 1; and

20 a compound having at least one cationically

polymerizable group in a molecule.

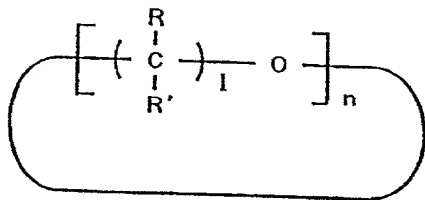
3. A photocurable composition as recited in claim 2, characterized in that said cationically polymerizable group is an epoxy group.

# A B S T R A C T

A cationic photocatalyst composition and a photocurable composition utilizing cationic polymerization are provided which can present sufficient open time and adhesion.

The cationic photocatalyst composition contains a photosensitive onium salt and a compound represented by the following formula (1). The photocurable composition contains the cationic photocatalyst composition and an epoxy compound having at least one epoxy group per molecule.

Formula (1) [Compound B]



(wherein R and R' are suitably selected from hydrogen, halogen, saturated hydrocarbon groups, unsaturated hydrocarbon groups, substituting groups comprising any suitable combination of elements such as carbon, hydrogen, oxygen, nitrogen and sulfur; and l and n each is an integer of 2 or larger).

F3715 PCT-US

ATTORNEY DOCKET NO: M&M-033-USA-PCT

### DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

CATIONIC PHOTOCATALYST COMPOSITION AND PHOTOCURABLE COMPOSITION

the specification of which is ☒ attached and/or ☐ was filed on ..... as Application Serial No. .... and was amended on (if applicable) .....  
☒ international (PCT) application No. PCT/JP99/04253 filed 05/08/1999 and as amended on ..... (if any).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

COUNTRY	APPLICATION NUMBER	DATE OF FILING	PRIORITY CLAIMED UNDER 35 U.S.C. 119
JAPAN	243175/1998	28/08/1998	<input checked="" type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

APPLICATION NUMBER	DATE OF FILING	STATUS (Patented, Pending, Abandoned)

I hereby appoint the following attorneys to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: Law Offices of Townsend & Banta: Donald E. Townsend, Registration No. 22,069; Teresa J. Banta, Registration No. 34,543; and Donald E. Townsend, Jr., Reg. No. 43,198  
Please address all correspondence to the Law Offices of Townsend & Banta, Suite 500, 1225 Eye Street, N.W., Washington, D.C. 20005

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

FULL NAME OF SOLE OR FIRST INVENTOR <u>Hiroji Fukui</u>		INVENTOR'S SIGNATURE <u>Hiroji Fukui</u>	DATE <u>November 15, 2000</u>
RESIDENCE <u>8-316, Hattanda-cho, Kisshoin, Minami-ku Kyoto-shi, Kyoto 601-8325 Japan JPX</u>		CITIZENSHIP <u>JAPAN</u>	
POST OFFICE ADDRESS <u>Same as Residence</u>			
FULL NAME OF SECOND JOINT INVENTOR, IF ANY		INVENTOR'S SIGNATURE	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			

Listing of Inventors Continued on Page 2 hereof. ☐ Yes ☒ No